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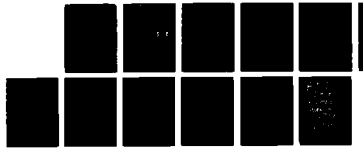

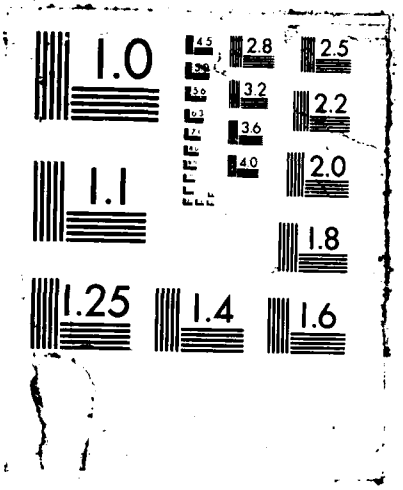
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by

S. G. Chung and Thomas F. George

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Generalized Levinson's Theorem and Quantum Sticking Coefficients at 0 K

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Abstract

Based on the generalized Levinson's theorem, some general properties of quantum sticking coefficients of an atom at 0 K are derived. Good agreement is obtained with experimental results for a ^{4}He atom colliding with a liquid ^{4}He surface.

The problem of quantum sticking coefficients α of an atom colliding with a solid surface has long been a matter of controversy. Lennard-Jones and Devonshire[1] first calculated α based on the distorted-wave Born approximation (DWBA), giving a vanishing coefficient at $T = 0$ K, or equivalently, when the wave number k of the incident atom goes to zero. The result, however, disagreed with the results of classical numerical calculations[2] and experiments both predicting unity instead of zero for sticking coefficients at $T \to 0$. As a possible explanation for such a disagreement between DWBA and experiments, Goodman[3] and Garcia and Ibanez[4] pointed out that a Morse potential[5] (exponentially decaying at large distance) used by Lennard-Jones and Devonshire for describing an adatom-surface interaction is unrealistic. They considered an algebraically decaying potential and obtained finite sticking coefficients. Their result, however, is inconclusive because the WKB approximation was erroneously used for the scattering states of the adatom. Other explanations have looked at correlated motions of the surface and the adatom. Krowles and Suhl[6] presented a polaron picture that when the adatom is near the surface, it becomes heavier due to virtual-phonon creations inside the matter. Although some enhancement was found for sticking coefficients, it still turned out that $\alpha \to 0$ as $k \to 0$. On the other hand, Doyen[7] considered self-trapping of an atom near a surface with a static distortion, obtaining the trivial results of $0 \leq \alpha \leq 1$. Although the self-trapping picture is in general more appropriate than the polaron picture for a short-range potential, Doyen's argument has a deficiency: Doyen assumed the existence of a resonant localized state for an atom near the surface as a quantum mechanical analog of the fact that in classical theory, the atom strikes the surface with probability of unity, but the existence or nonexistence of such a state in quantum theory is the very core of the issue.

In this paper, we develop a new theory of quantum sticking coefficients at T = 0 K. We have recently generalized Levinson's theorem to an atom dynamically interacting with a one-dimensional (1-d) matter system.[8] Based on the theorem, we derive here the following properties of α at T = 0 K: the small-k dependence of $\alpha(k)$ is determined by the long-distance behavior of adatom-surface potentials. For potentials decaying faster than x^{-2}, where x is the adatom-surface separation $\alpha(k) \propto k$ for small k. For potentials decaying as x^{-2} or slower, $\alpha(0)$ can be finite. The theory encourages experiments on α at small k $\lesssim 0.1$ Å^{-1}. For a ^{4}He atom colliding with a liquid ^{4}He surface, the present theory predicts a critical wave number ~ 0.01 Å^{-1} below which $\alpha(k)$ depends on k approximately linearly, which is in good agreement with experiment.[9]

Let us consider the adatom + 1-d matter Hamiltonian

$$H_{tot} = H(\tilde{x},\tilde{p}) + V(\tilde{x},x) + K(p) \quad , \tag{1}$$

where $H(\tilde{x},\tilde{p})$ is a 1-d matter Hamiltonian with $\tilde{x}$ and $\tilde{p}$, respectively, representing position and momentum vectors for atoms in the 1-d matter, $V(\tilde{x},x)$ is an interaction between the adatom and 1-d matter, and K(p) is the kinetic energy of adatom. A scattering eigenstate of (1) with a wave number k and a boundary condition that $\psi(\tilde{r}_c,k) = 0$, where $\tilde{r}_c$ is an appropriate constant vector independent of k, can be written as

$$\psi(\tilde{r},k) = \frac{i}{2k}\,[f(-k)F(\tilde{r},k) - f(k)F(\tilde{r},-k)] \quad . \tag{2}$$

Here $\tilde{r} \equiv (\tilde{x},x)$, $f(k) \equiv F(\tilde{r}_c,k)$ is a Jost function, and $F(\tilde{r},k)$ is a Jost solution having an asymptotic form $e^{-ikx}\phi_0(\tilde{x})$ as $x \to \infty$, where $\phi_0(\tilde{x})$ is the ground state of $H(\tilde{x},\tilde{p})$ (T = 0 K).

We have recently shown that Levinson's theorem[10] in static potential scattering applies to our case in a similar manner:[8]

$$N_b = \delta(+0)/\pi \quad , \tag{3}$$

where N_b is the number of bound states of H_{tot}, and $\delta(k)$ is a scattering phase shift of the adatom.

Based on the generalized Levinson's theorem (3), we now derive some general properties of quantum sticking coefficients $\alpha(k)$, at T = 0 K. From time-dependent scattering theory,[11] one can write $\alpha(k)$ as

$$\alpha(k) = \frac{2\pi M_a}{\hbar^2 k} \sum_f \delta(E_k - E_f) \; |\langle f|V_f|k^+\rangle|^2 \quad , \tag{4}$$

where $\sum_f$ means a summation over final states $|f\rangle$, V_f represents a final channel interaction, e.g., creation of phonons, and $|k^+\rangle$ is the scattering eigenstate (2) with a trivial change of normalization. Let us consider $k \ll 1$ such that the S-matrix element $S(k) \equiv e^{2i\delta(k)} \sim 1$, and a large distance $x = R$ such that

$$-U(R) \equiv -V(\tilde{x}_0, R)/(\hbar^2/2M_a) = k^2 \quad , \tag{5}$$

where $\tilde{x}_0$ represents the mean position of the matter when the adatom-matter distance is large. Let us assume, for the moment, an inequality

$$kR < 1 \quad . \tag{6}$$

Under these conditions we have for $R < x < 1/k$

$$|k^+\rangle \cong -2ik\phi_0(\bar{x}) \quad . \tag{7}$$

We now claim that once (7) is realized, then for $0 < x < R$

$$|k^+\rangle \propto k \quad , \tag{8}$$

where $x = 0$ denotes the surface. This can be seen as follows: over the region $R' < x < R$, where R' is such that $k^2 << -U(R') << (-U(x))_{max}$, $|k^+\rangle$ smoothly changes from its behavior at $x \sim R'$ to its behavior at $x \sim R$. The behavior of $|k^+\rangle$ in this region must be somewhere between the following two extreme cases: (a) $U(x) = U(R)$ and (b) $U(x) = U(R')$. But one can easily see that in both cases the requirement of smoothness of $|k^+\rangle$ at $x = R$ leads to $|k^+\rangle \propto k$ at $R' < x < R$. This fact, in turn, leads to $|k^+\rangle \propto k$ at $0 < x < R'$, because in this region $|k^+\rangle$ can depend on k only through a constant prefactor which, however, again by smoothness of $|k^+\rangle$ at $x = R'$, must be proportional to k. In this way we reach (8).

From (4) and (8) we have

$$\alpha(k) \propto k \quad . \tag{9}$$

We remember, however, that (9) is derived under the condition (6) and $S(k) \sim 1$. Little is known about the small-k dependence of S(k). Our analysis and numerical calculations below for a Morse potential (see Fig. 1) show that the latter condition imposes a stronger restriction on k than (6). We now analyze (6) for typical long-distance behaviors of the potential U(x). Let us introduce a standard number $\beta < 1$ in analogy to the "standard value" in the Lindemann melting formula.[12] With this β we can predict for each given potential a critical wave

number k_c below which $\alpha(k)$ depends on k linearly (approximately when the condition $S(k) \sim 1$ is not fulfilled). We have obtained the following results:

(i) For $U(x) \sim -\gamma e^{-\kappa x}$, $\alpha(k) \propto k$ at $k \leq k_c$,
where $-(k_c/\kappa)\ln(k_c^2/\gamma) = \beta$. (10a)

(ii) For $U(x) \sim -\gamma x^{-n}$ $(n > 2)$, $\alpha(k) \propto k$ at $k \leq k_c$,
where $\gamma^{1/n} k_c^{1-2/n} = \beta$. (10b)

(iii) For $(Ux) \sim -\gamma x^{-n}$ $(n \leq 2)$, $k_c = 0$, which strongly indicates a possibility of finite $\alpha(0)$. (10c)

Numerical calculations below for a Morse potential (10a) with $\gamma = 48$ Å^{-2} and $\kappa = 1.4$ Å^{-1} (numbers for the W-He system) show that $k_c \sim 0.1$ Å^{-1}, which leads to $\beta \sim 0.6$. The ^{4}He atom colliding on a liquid ^{4}He surface is described by a van der Waals potential (10b) with $n = 3$ and $\gamma = 20$ Å. Using $\beta \sim 0.6$ we have $k_c \sim 0.01$ Å^{-1}, in good agreement with experiment.[9]

We have perfomed a numerical calculation on a simple self-trapping model. The model assumes a harmonic lattice, $H_{ph} = \sum \hbar\omega_q a_q^\dagger a_q$, a surface atom plus an adatom interacting through a Morse potential, and the surface atom seeing another Morse potential created by the end atom of the 1-d lattice in its equilibrium (taken to be $x = 0$),

$$H_{s+a} = P_s^2/2M_s + \bar{U}(x_s) + P_a^2/2M_a + U(x_a - x_s) \quad . \tag{11}$$

The dynamical interaction between the harmonic lattice and the surface atom (one-phonon process) takes the form

$$H_{int} = -\sum_{q} \left(\frac{\hbar}{2N_\ell M_s \omega_q}\right)^{\frac{1}{2}} (a_q^\dagger + a_{-q}) \frac{dU(x_s)}{dx_s} \Theta(t) \quad . \tag{12}$$

In (12), N_ℓ is the number of lattice atoms, and the unit step function $\Theta(t)$ means that $|k^+\rangle$ here is a scattering eigenstate of H_{s+a}. In analogy to the second-order optical process, this is a "luminescence component" approximation thereby neglecting "Raman components", which is expected to be good for treating short-range potentials of the Morse type. We note that V_f in (4) is H_{int} at $t > 0$. Thus our main task is to solve the Schrödinger equation $H_{s+a}\psi = E\psi$.

As an example, we have considered W-W and W-He Morse potentials:

$$\begin{pmatrix} \tilde{U}(x+x_0^s) \\ U(x+x_0^a) \end{pmatrix} = \begin{pmatrix} \tilde{D} \\ D \end{pmatrix} (e^{-2\kappa x} - 2e^{-\kappa x}) \quad , \tag{13}$$

where $x_0^s \sim 3$ Å, $x_0^a \sim 3.6$ Å, $\tilde{D} \sim 1$ eV, $D \sim 10$ meV and $\kappa = 1.4$ Å^{-1}.[5,13] The one-body problem for Morse potentials is exactly solvable:[5] the W-He potential $U(x)$ supports 4 bound states, whereas the W-W potential $\tilde{U}(x)$ supports about 200, and the energy separations of the two adjacent lowest bound states are about 10 meV. With these facts in mind, we consider following variational wave functions for the eigenstates of H_{s+a}:

$$\psi(x_s,x_a) = C\phi_0(x_s)\,(e^{-ikx_a} - Se^{ikx_a})\,\phi(x_a)$$

$$+ \sum_{p=0}^{2} \sum_{q=0}^{N-1} C_{pq}\, \phi_p(x_s)\, h_q(x_a) \quad , \tag{14}$$

where C = 0 for bound states and C = 1 for $|k^+\rangle$. Three ϕ_p's are confluent hypergeometric functions representing the lowest 3 bound states of the surface W atom.[5] $\Phi(x_a)$ is an error function preventing the adatom from penetrating into the solid. Finally, the h_q's are Gaussian weighted Hermite polynomials,[14] which are found to be best suited for describing fully correlated motions of the He atom near the surface.

The Schrödinger equation is thus converted to a set of coupled algebraic equations for the coefficients S and C_{pq}'s. The stability and convergence of the numerical calculation has been checked by changing a certain set of variational parameters as well as the number N of h_q's. Figure 1 contains our final results for N = 21. Part a shows $|S|$ and ReS as functions of the wave number k, demonstrating the generalized Levinson's theorem (Eq. (3)). Part b shows $\alpha(k)$ vs. k. The approximate linear dependence of α on k is seen below $k_c \sim 0.1$ Å^{-1}.

To summarize, we have generalized the Levinson's theorem to an atom dynamically interacting with a 1-d matter, and based on this we have derived some general properties of quantum sticking coefficients at T = 0 K. A generality of the present theory strongly encourages further accurate experiments on sticking coefficients in a variety of physical systems. Theoretically, on the other hand, we must determine if $\alpha(0)$ is finite in the case $U(x) \sim x^{-2}$ at large x. The result of this study along with details of the present paper will be reported elsewhere.

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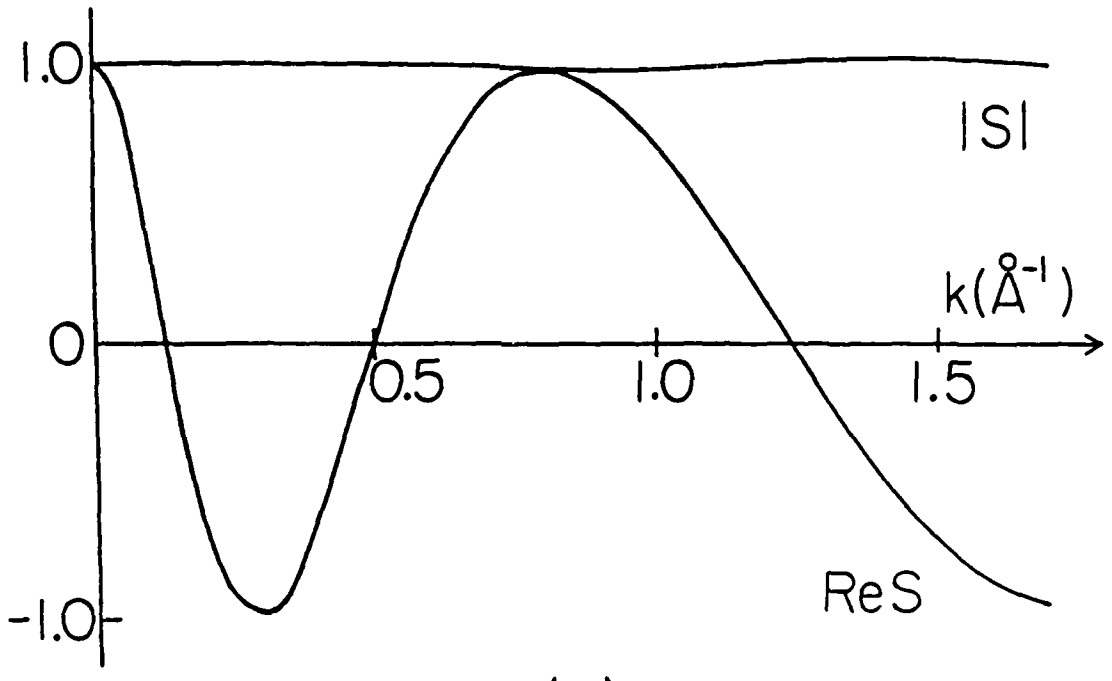
Figure Caption

Fig. 1 (a) $|S|$ and ReS vs. K. It is seen that $S \to 1$ as $k \to 0$. (b) $\alpha(k)$ vs. k. Below $k_c \sim 0.1$ Å^{-1}, α has an approximate linear dependence on k.



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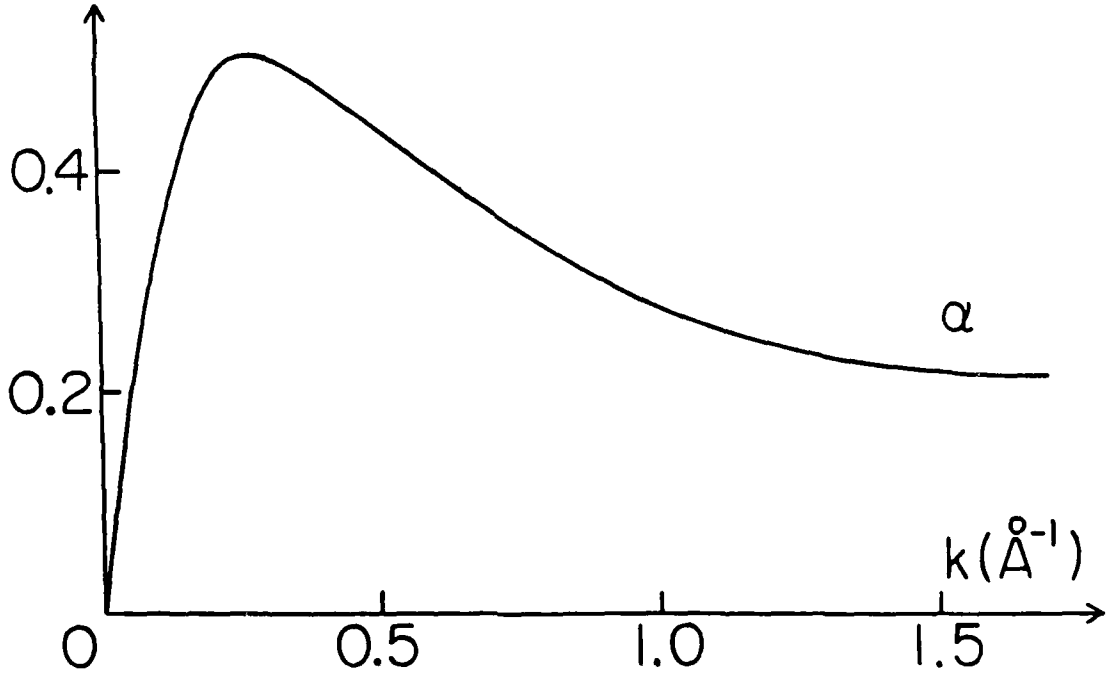

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Fig. 1

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